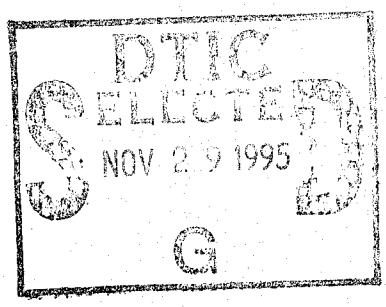
Atmospheric Radiation Measurement Program (ARM)— Summer 1995 Review



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Atmospheric Radiation Measurement Program (ARM)— Summer 1995 Review

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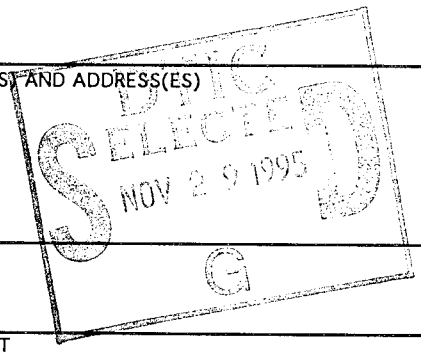

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This report examines the issues of anomalous atmospheric absorption and makes recommendations concerning future directions for the ARM program.

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Contents

1 ATMOSPHERIC RADIATION MEASUREMENT PROGRAM (ARM) — SUMMER 1995 REVIEW

ARM is a highly focused program designed to improve our understanding of the transport of infrared and solar radiation through the atmosphere. The program pays particular attention to the interaction of radiation with the three phases of water. The goals of ARM are usually articulated in terms of improvements in climate models. We agree that ARM can indeed make significant contributions to the understanding of climate change. In addition we believe that the results of the program will have wide applicability to a broad range of problems, including more accurate short-term and seasonal weather forecasting.

ARM has made remarkable progress over the past two years. The Southern Great Plains (SGP) site is operational, though further instrumentation will increase its value. The ARM data system is delivering high-quality data to its science team. This system includes data not only from the ARM array of instruments, but also from the NOAA radar and wind profiles, operational satellites, and the Oklahoma Mesoscale Measurement Network. The ARM archive is storing this data in a format that makes for easy retrieval by scientists worldwide.

Preparations for the second site in the Tropical Western Pacific (TWP) have made significant progress, and the site should be operational in 1996. The TWP site will contribute to the understanding of such phenomena as El Niño, and therefore to the possibility of seasonal forecasts. Preliminary

work is also underway to prepare for an ARM site on the North Slope of Alaska, which will aid in understanding atmosphere/radiation interactions in the Arctic.

The Department of Energy's ARM program is clearly the jewel of the U.S. climate change program. ARM is well managed, and has gained the attention of many of the world's leading atmospheric scientists. Strong institutional ties have been established between the program and government agencies such as NOAA and NASA, and between the program and universities and international groups

Against this glowing picture of ARM, we are increasingly concerned about the program's future—a concern that is increased by our recognition of ARM's potentially great value in several areas. Our uneasiness centers on the linked issues of central mission, budget, and wide use of the ARM facilities by other groups.

Documents describing ARM, and public presentations on ARM, invariably emphasize ARM's contribution to improving global circulation models (GCMs), and thus to deepening our understanding of global climate change. As noted above, we believe that ARM does indeed play a key role in climate change research. However, this narrow definition of ARM's mission should not hide the fact that understanding radiative transport in the atmosphere is also central to improving day-to-day weather forecasting. Support for climate change research is highly vulnerable to shifts in political winds, but no one questions the economic value of better weather prediction. We understand that weather forecasting is in NOAA's domain, but strong collaboration between NOAA and ARM can dampen this potential turf skirmish. Turf concerns cannot be allowed to suppress the recognition that ARM will

make significant contributions both to weather forecasting at the time scale of days and, through the TWP-like site, to seasonal forecasts.

At best, ARM can anticipate level funding over the next few years. Overall budget pressures indicate that the ARM budget may very well be cut. Already, the SERDP support for the UAV component of ARM has melted away. In this stringent budget situation, there is danger that the program will spend in such a way that the quality and depth of ongoing activities will suffer. We would give the SGP site the highest priority, with the TWP site a strong second priority. A strong case can be and has been made for an Arctic site, work on this site should not proceed if doing so would imperil programs underway at the SGP and TWP sites.

DOE funding has created a unique facility at the SGP site. An array of instrumentation provides the capability to describe the state of the atmosphere with a greater resolution and fidelity than is available at any other site. At the same time, interest in remote sensing is exploding worldwide. NASA is poised to launch its Earth Observing System. Three U.S. commercial consortia have obtained licenses to place in orbit high-resolution (1-meter) photographic systems. The U.S. classified community continues a very active, advanced program in remote sensing, as do France, China, India, Brazil, and Canada, among others. Commercial firms worldwide carry out airborne remote sensing operations. A significant requirement in all these activities is the validation of the remotely sensed data. In many cases, instrument calibration requires a well characterized atmosphere. The SGP site meets many of the requirements of a validation/calibration site for remote sensing instrumentation.

As a result of a JASON suggestion, DOE is currently negotiating with the classified imagery community to use the ARM site for validation and calibration. In addition, ARM and NASA have begun discussions with respect to using the SGP site for validation. We believe that ARM management should aggressively pursue further possibilities for use of ARM facilities by other developers and users of remote sensing technology. In fact, we recommend that ARM management prepare a "business plan" for the use of the SGP site by government and commercial users of remote sensing. Such activities will increase the visibility and support, both political and budgetary, of ARM activities. Given that political support for climate change-oriented ARM programs may waver in the future, an ARM program with multiple goals will have a better chance for long-term, stable support.

2 RADIATIVE ENERGY BUDGETS IN THE ATMOSPHERE

Solar energy deposited in the atmosphere is a major driver of climate: globally, about 20 percent of the radiant energy from the sun is directly absorbed within the atmosphere. A study of radiative transport in the atmosphere divides into several distinct parts: (1) the relevant microphysics-elastic and inelastic cross-sections for scattering of photons from atmospherically relevant targets (molecules, molecular clusters, particulates, raindrops, ice crystals); (2) given the microphysics, the development of codes that can "follow the photons" as they multiply scatter and absorb in a broad range of physically relevant model atmospheres; and (3) applications to actual or climate model atmospheric states characterized by altitude, geographic location, time of day, water content, cloud cover and structure, aerosol concentrations, etc. In the real world, (2) and (3) are, of course, coupled. The state of the atmosphere is in part determined by radiative energy flow, while the latter depends in part on the state of the atmosphere.

The propagation and absorption of radiation in a specified atmosphere, if all the relevant cross-sections are given, constitute the easiest part of the problem, at least conceptually. The governing equations are beyond much doubt. Computationally, however, they are very demanding, so that approximations and truncations must be adopted. These are incorporated in elaborate codes that have developed over time and that are now widely used. For application to a real situation, one has then to specify appropriately the relevant atmospheric parameters. Radiative transport experiments carried out in the real atmosphere can have two kinds of objectives: to learn about

the atmosphere, accepting that the theoretical model is sufficiently correct, or to test the model (and the microphysics underlying it) supposing that the atmosphere is sufficiently well specified. Inevitably, there is some of both in any experiment.

When clouds are present, adequate characterization of the atmosphere being probed experimentally can be very problematic. A considerable phenomenology of cloud physics has been developed over the years, and many of the characteristics that are relevant for radiative transfer have been clearly identified: cloud dimensions and cloud column water content, drop size, distribution, etc. But these parameters are not always fully measured for the particular cloud system that enters into a given experiment. Moreover, the gross geometric parameters (thickness, area) do not do justice to cloud inhomogeneities, holes, reflections off the sides of cloud patches, etc. Clouds are complicated and variable objects. For clear-sky experiments, the situation is certainly easier; but here, too, important parameters such as the aerosol content and size distribution in the particular patch of sky being studied are not always easy to measure.

All this having been said, a considerable stir has been created by recent report of substantial discrepancies between theory and experiment concerning atmospheric absorption in the solar (the shortwave) band from .3 to 2.5 μm. Especially significant is the claim of substantial excess absorption in clear-sky conditions. Excess absorption by clouds is similarly reported. The experiments are summarized below.

Two attempts have been made to determine by direct observations the excess solar absorption by clouds. The two experiments are similar in concept, but differ significantly in detail. Pilewskie and Valero (1995) flew two

airplanes below and above clouds. The two airplanes carried very well-calibrated radiometers responsive in the shortwave band. The net flux—downwelling minus upwelling—was measured nearly simultaneously below and above the clouds. They found that the ratio of cloud forcing at 20 km to that at the surface is 1.58, rather than 1.0 as predicted by radiative transfer models. However, the measurements were on total flux over the entire shortwave band.

Hayasaka, Kikuchi, and Tanaka (1995) carried out similar flights over and under strato-cumulus clouds. The aircraft were equipped with a pair of pyrometers and near-infrared pyrometers. Downward and upward shortwave fluxes below and above the clouds were simultaneously measured by the two airplanes, as in the case of the Pilewskie and Valero experiments. In the Japanese experiment, the visible shortwave radiation was subtracted from the total net radiation to obtain the shortwave near-infrared absorption. The interpretation of data then followed by assuming that the scattering for both the visible and near-infrared was similar, but that the absorption took place only in the near-infrared. Measurements of the vertical convergence of visible radiation (measurement of the visible flux at the top and bottom of an atmospheric column) becomes by continuity a measurement of the horizontal convergence, since, by assumption, there is no absorption in the visible. Any vertical convergence of the visible must be balanced by horizontal divergence.

Hayasaka et al. (1995) obtained good agreement with expected radiative absorption, pointing out that the apparent excess absorption resulted from the horizontal divergence and convergence that in turn resulted from multiple scattering in the inhomogeneous cloud.

Both aircraft experiments were carefully carried out with well-calibrated instruments. There is an urgent need to determine whether the excess absorption observed by Pilewskie and Valero correctly assesses conditions. The proposed set of observations to be carried out at the Oklahoma ARM site in September 1995 are essential to determining the real situation. We urge that these experiments attempt to include observations of the horizontal flux of radiation in the vicinity of clouds, using either the spectral technique employed by Hayasaka et al. or direct observation.

Li et al. (1995) have discussed the problem of possible excess absorption. They emphasize the possible contribution of aerosols, particularly in tropical regions (see Section 4). Clearly, the ARM experiments should include a detailed characterization of the atmospheric aerosols and their vertical distribution.

On the basis of current experimental data, we have a number of observations. For ideally clear skies with very small aerosol content and low humidity, the principal absorbers must be water molecules and the water column can presumably be well measured. It would thus have to be the fundamental water cross-sections, in particular, the shape of the absorptive resonant curves, that come into question to account for any discrepancies (see Section 3). The basic microscopic information is in any case important and worth experimentally checking and improving. In particular, it is now possible in the laboratory to obtain very long path-length measurements, and a variety of techniques could be employed, including using the photoacoustic effect. A small laboratory program in support of the field observations appears essential. For many years, the radiative transfer models have been developed by the Air Force's Cambridge Geophysical Laboratory, and the codes have been maintained by this group. If there are errors in these codes,

there would be significant repercussions in a whole variety of activities.

As noted in Section 1, the ARM program is well situated to make contributions of very great importance to many aspects of atmospheric sciences. We encourage the program's efforts to further understand cloud absorption. At the same time, we note that a number of techniques, including bubble chamber techniques, could have wide applicability to measuring water vapor absorption at high relative humidities now considered difficult because of condensation on the walls. There is room for much progress in both the laboratory and in the field, and such progress is essential if we are to understand radiative transfer in the atmosphere more clearly.

3 MODELING SHORT WAVE ABSORPTION BY WATER VAPOR

In principle, the absorption of optical and near IR solar radiation by water vapor can be measured in the laboratory at the partial pressures and temperatures which are relevant in clouds or "clear air". However, such measurements are made difficult by enormous differences between the geometrical scales.

(a) Relevant atmospheric path lengths are typically up to 10^5 times longer than the sizes of convenient laboratory chambers. An absorption could be so small that it is hard to determine in the laboratory, but it may still be very significant over the much longer path lengths in a slightly absorbing atmosphere. We note, however, that techniques for multi-reflected paths are continually becoming more sophisticated so that effective absorbing lengths in laboratory experiments can be made to exceed hugely the length scale of laboratory chambers. Even more important may be the application of photoacoustic techniques to detect very minute absorption from short laser pulses. Absorption as small as 10^{-10} of the incident flux can be measured in this way: sufficiently small to permit results in the laboratory to be directly applied to atmospheric scales.

(b) In clouds, water vapor is generally slightly supersaturated, and often vapor in clear air may be near this regime. In laboratory chambers, however, it has proved difficult to measure absorption in water vapor when the relative humidity exceeds 80%: above

this humidity vapor condenses on windows (and walls), which compromises vapor absorption measurements. [A similar problem was successfully handled many decades ago when cloud chambers were commonly used in high energy experimental physics to make visible the ionizing tracks of energetic charged particles. The chamber gas was supersaturated, but non-track-associated condensation was avoided because the chamber gas was supersaturated only very briefly (less than a second) during each sudden expansion of the chamber when observations were made. Window heating could be used where needed. Steady state ways of avoiding condensation on chamber surfaces for metallic vapors also seem directly applicable to water vapor.]

(c) To maintain the large water vapor densities in laboratory experiments currently needed to measure absorption in weakly absorbing spectral regions, the vapor temperature is generally kept very much higher than that found several kilometers up in the atmosphere. (In some cases T(lab) = 353 K, about 100 degrees higher than the atmospheric one.)

In the absence of available sensitive measurements over all relevant spectral regions in which the laboratory vapor may be quite transparent but many kilometers of nearly saturated vapor may not be, theoretical models are needed to compute accurately the water vapor contribution to solar absorption. They must be used to extrapolate from the large water vapor densities in many experiments to the much smaller ones appropriate to various atmosphere altitudes.

The vibrational and rotational frequencies of an isolated water molecule are well established. The integrated absorption (over frequency) of each spectral line is also well determined by sum rules for any given probability distribution among the vibrational and rotational states of the molecules. Difficulties in predicting the detailed absorption spectra arise from two related uncertainties about the dynamics of colliding H_2O molecules (with N_2, O_2 and H_2O) in a real atmosphere.

(a) What is the line shape? If relaxation and excitation collisions (among rotational states) were all instantaneous and the vibration and rotation frequencies of the molecule did not vary during its close approach to other molecules, each line would have the well-known normalized Lorentz shape

$$f(\omega) = \frac{\alpha/\pi}{(\omega - \omega_0)^2 + \alpha^2} ,$$

where α is the effective excitation/deexcitation collision rate and ω is the spectral angular frequency. In this idealization the distant wing of each absorption line falls off like $(\omega - \omega_0)^{-2}$, and extrapolation to spectral regions far from absorption line centers would be relatively straightforward. The parameter α could be measured in the more strongly absorbing regions. However, the more distant line wings are sensitive to details of the collision as soon as $|\omega - \omega_0| \gtrsim \hat{\tau}_c^{-1}$ (the inverse of the collision duration $(\hat{\tau}_c)$). If there is no complication from transient dimer formation, $\hat{\tau}_c \sim a/v$, with a the effective interaction distance for a collision and v the effective relative velocity for the collision (adjusted for the strong dipole-dipole interaction in H_2O-H_2O collisions). For $a \sim 10^{-8}$ cm and $v \sim 5 \cdot 10^4$ cm s^{-1}, $\hat{\tau}_c \sim 2 \cdot 10^{-13}$ s and the line width beyond which the idealized Lorentz line shape is expected

to fail is $\Delta(1/\lambda) \sim 25$ cm^{-1}, of order but still considerably less than the separation between H_2O's rotational lines. [By comparison, the α of the canonical Lorentz form for pure water vapor at a vapor density $\sim 3 \cdot 10^{19}$ cm^{-3} (one atmosphere) corresponds to $\Delta(1/\lambda) \sim 0.5$ cm^{-1}.]

In water vapor absorption modeling (e.g., Clough et al. (year)) the spectral lines are appropriately taken as Lorenztian for $\Delta(1/\lambda) <$ 25 cm^{-1} and the line shape (arbitrarily) adjusted to fit available measured data further from the line centers. It is found, quite plausibly, that the most distant parts of Lorentz line wings are suppressed (because of the absence of suddenness in real collisions) while the less distant parts are increased. ($\int_{-\infty}^{\infty} f(\omega)d\omega$ must remain unchanged.)

(b) Is there transient H_2O clustering (dimers or polymers) at the low temperatures of the atmosphere many kilometers above the earth's surface, for which vibrational frequencies are significantly shifted and greatly broadened? If so, there can be contributions to absorption very far from the absorption lines of unperturbed solitary H_2O molecules that would not necessarily be reflected in the "semi-empirical" parameterization of line shapes, which may be adequate much nearer to those lines.

The "bottom line" is that experimental verification of present theoretical modeling seems needed where such models are to be extended to more weakly absorbing spectral regions of water vapor in the real atmosphere. Such regions exist between rotational lines of a vibration band and also far from the vibrational band center. Where direct data are absent, deficiencies may exist in the models because they lack detailed quantitative descriptions of all of the

possibly relevant dynamics in H_2O collisions. As such verification becomes available, a model is needed only to extrapolate results to regimes with different densities and temperatures (which probably affect mainly collision frequencies but would be much more important if dimer formation is significant). This is generally done on the assumption that clustering does not contribute and can then be accomplished simply and reliably. In spectral regions where direct measurements do not yet exist and predictions depend on what can be overly simple parameterizations of collision dynamics, verification is needed. It would seem of high priority, therefore, to devote resources to exploit-state-of-the-art techniques to improve laboratory experiments on water vapor absorption. This can greatly reduce dependence on quantitatively uncertain theoretical extrapolations.

4 AEROSOL MEASUREMENTS

An adequate quantitative description of the aerosol content of "clear air" and of clouds, and of the dynamical role of aerosols in determining cloud structure, may be crucial for interpreting much climate data, for constructing reliable GCMs, and for comparing climate models with observations.

A major fraction of the aerosol in our present atmosphere consists of sulfates whose origin is anthropogenic sulfur emission from coal burning and smelting. This emission has grown enormously over the past century. By 1910 it exceeded natural sulfur emission in the Northern Hemisphere; it is now about 5 times larger. Sulfur emission affects the partition of incident sunlight in at least two ways. After oxidation and the formation of sulfate aerosol particles (probably mainly the residues of evaporated or nearly evaporated sulfate nucleated cloud droplets), aerosol backscattering of incident sunlight contributes some albedo even to nominally "clear" air. Even if sulfate aerosol had no other effects, this increased "clear air" albedo is estimated to cool the surface of the earth by an amount quite similar in magnitude to the increase in surface temperature that GCMs indicate would otherwise have resulted during the past 80 years from increasing CO_2 emission. (The estimated counterbalancing of surface temperature changes from CO_2-induced greenhouse warming and "whitehouse cooling" from anthropogenic aerosols will probably soon fail: the rate of aerosol formation is no longer increasing as sulfur control measurements are instituted, while CO_2 emission continues to rise.)

It is much more difficult to calculate the indirect effects of the sulfur emission on cloud albedo. That fraction of emitted sulfur which is oxidized to sulfate before it is dissolved in a cloud droplet can form sulfate particle

aggregates which increase the number density of cloud condensation nuclei (CCNs). This may have little consequence where there is already an abundance of CCNs, e.g., near the SGP ARM site. However, at sites which are at present far removed from existing anthropogenic sources for CCNs, the impact of increased sulfur from anthropogenic or biological sources may affect cloud droplet number density, droplet size, and lifetime. Such changes would alter cloud albedo. A cloud albedo change of only a few percent could impact climate as greatly as changes estimated from increased future CO_2 emissions. It would seem necessary, therefore, to measure and understand how existing aerosols affect "clear" air albedo measurements, sunlight absorption (mainly from carbonaceous aerosols), and cloud properties around both industrially active areas (e.g., near the SGP ARM site) and more pristine oceanic ones (e.g., the Tropical Western Pacific). Of special interest at present is determining possible aerosol contributions to reported "anomalies" in cloud and in "clear air" shortwave absorption. A clear air "absorption" anomaly as large as 40 wm^{-2} (at noon) has been suggested by some experiments. This is perhaps, but not certainly, an order of magnitude larger than would be accounted for in models which include aerosols. Results may vary considerably depending upon the amount of water that hydroscopic sulfate particles had accumulated when the measurements were made.

Ultimately it will be necessary to evaluate quantitatively a number of coupled relationships whose combined effect plays a role in determining how greenhouse-induced changes feed back to change cloud and clear air albedo:

(a) How would the emission of (reduced) sulfur compounds by ocean biota vary with possible future changes in ocean surface temperature and flow?

(b) How would clear air shortwave albedo above the oceans respond to such changes?

(c) How would the changes in available atmospheric sulfate and CCNs impact the fraction of clear air to cloud cover?

(d) How would it change cloud albedo?

It does not seem premature to begin to give more emphasis to measurements of correlations between both cloud and clear air effects on incident solar radiation and their sulfate content and its aggregation.

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